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Accurate mass measurements on neutron-deficient krypton isotopes

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Abstract

The masses of $^{72-78,80,82,86}\text{Kr}$ were measured directly with the ISOLTRAP Penning trap mass spectrometer at ISOLDE/CERN. For all these nuclides, the measurements yielded mass uncertainties below 10 keV. The ISOLTRAP mass values for $^{72-75}\text{Kr}$ outweighed previous results obtained by means of other techniques, and thus completely determine the new values in the Atomic-Mass Evaluation. Besides the interest of these masses for nuclear astrophysics, nuclear structure studies, and Standard Model tests, these results constitute a valuable and accurate input to improve mass models. In this paper, we present the mass measurements and discuss the mass evaluation for these Kr isotopes.

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I. INTRODUCTION

Accurate atomic mass measurements are essential in several branches of physics. As a consequence of the mass–energy equivalence, the mass of an atom provides a direct determination of the nuclear binding energy. Thus, systematic mass measurements are relevant for testing and further improving nuclear mass models. In the case of exotic nuclides, specifically of those close to the proton drip-line, accurate mass measurements are essential for astrophysics and weak interaction studies including tests of the Standard Model (SM) [1–3].

In astrophysics, the atomic mass of the so-called “waiting points” must be known with a precision of 10^{-7} or better in order to expand reliable nucleosynthesis calculations together with the rp -capture process [1]. The rp -process takes place, among several other sites, in Type I X-ray bursts [1, 4, 5]. There, waiting points cause a delay of the process, hampering the formation of heavier elements. The knowledge of proton separation energies through accurate masses of the nuclei involved can yield an accurate determination of this delay and therefore the range of the process at high Z . As a result of the measurements reported here (specifically on $^{72-74}\text{Kr}$), the effective lifetime of the $N = Z$ nuclide ^{72}Kr ($T_{1/2} = 17.2$ s) was shown to be very close to its β -decay lifetime. This confirmed that ^{72}Kr is a strong waiting point in the rp -process [6].

An even more precise knowledge of the mass is required to carry out tests of the SM, specifically to validate at high Z the Conserved-Vector-Current (CVC) hypothesis and to test the unitarity of the Cabibbo-Kobayashi-Maskawa (CKM) quark mixing matrix [2]. For these tests, the required mass value accuracy is 10^{-8} and must be reached in some cases for very short-lived nuclei. The binding energy Q from mass measurements can be used to determine the statistical-rate function, or F -value, which is a function of Q^5 [7]. For a particular superallowed transition, the Q -value is obtained from the mass difference of the parent and the daughter nuclei.

Regarding the SM tests, the measurements on ^{74}Kr , in combination with the measurements on ^{74}Rb [8], have provided the first meaningful comparative half-life, or Ft -value, for nuclei with $A > 54$ [9]. This quantity is determined from the Q -value, half-life, branching ratio and the calculated electron capture fraction in the decay. The determination of Ft also requires some corrections, which account for the fact that the decay takes place within the nucleus. These corrections are the isospin-symmetry-breaking parameter δ_C and the ra-

diative correction parameter δ_R [3]. In the case of the decay $^{74}\text{Rb}(\beta^+)^{74}\text{Kr}$, the uncertainty of Ft is now limited by the incomplete knowledge of δ_C [9]. However, assuming the CVC hypothesis holds, the ISOLTRAP results on ^{74}Kr and ^{74}Rb is used to obtain an experimental value for this correction parameter at high Z . This constitutes a valuable input for further theoretical work.

All these issues called for an accurate determination of the mass of neutron-deficient krypton isotopes using Penning trap techniques as provided by ISOLTRAP. ISOLTRAP provides the required accuracy on the nuclei of interest. The use of any mass value must be preceded by a proof of reliability of the technique employed for the measurement. This becomes more important in those cases where the accuracy is pushed down to the limits mentioned above. In the course of the first ISOLTRAP experiment on Kr isotopes, discrepancies were observed between the ISOLTRAP mass values for $^{73-76,78}\text{Kr}$ and the results obtained by means of other (indirect) techniques. Due to these discrepancies, a second experiment was scheduled to measure again the masses of $^{73,74,78}\text{Kr}$, and to determine the mass of ^{72}Kr for the first time. While the earlier ISOLTRAP values could be confirmed, some of the discrepancies remained and will be discussed in this paper.

II. EXPERIMENTAL SETUP

The ISOLTRAP mass spectrometer [10–12] is located at the ISOLDE facility [13] at CERN/Geneva (Switzerland). A pulsed proton beam with 1.4 GeV energy at a repetition rate of 1.2 s and average intensity of $2\ \mu\text{A}$ hits a thick target. The reaction products diffuse out of the target, heated to temperatures of up to 2600 K, into the ion source. The atoms are ionized either by using laser ionization, surface ionization, or hot plasma ionization. The ion beam is then accelerated to 60 keV and mass separated in one of two mass separators. The General Purpose Separator (GPS) and the High Resolution Separator (HRS) operate usually with mass resolving powers $m/\Delta m \approx 1200$ and 6000, respectively. Finally, the mass-separated ion beam is delivered to different experimental setups.

The ISOLTRAP mass spectrometer is shown in Fig. 1. It is comprised of three different traps: A gas-filled linear Paul trap [11], a gas-filled cylindrical Penning trap [14] and a hyperbolic precision Penning trap in high vacuum [10].

The 60-keV ISOLDE beam with an energy spread of a few eV and an emittance of

about $30\pi\cdot\text{mm}\cdot\text{mrad}$ (after the HRS) is electrostatically retarded to about several ten eV and stopped in the gas-filled linear Paul trap. There the ions are cooled by collisions with ≈ 0.5 Pa helium buffer gas. After an accumulation time which was varied depending on the production yield of the isotope to be investigated, the cooled ion bunch is ejected with a temporal width of less than $1\ \mu\text{s}$ and an emittance of less than $10\pi\cdot\text{mm}\cdot\text{mrad}$.

The ion bunches are transported to and captured in the helium buffer-gas-filled cylindrical purification Penning trap. This trap allows for isobaric cleaning of the injected ion bunch [15]. A resolving power up to 10^5 has been reached with a trapping time of 500 ms at mass number $A = 138$ and 10^{-3} Pa helium buffer gas [14]. After the isobaric cleaning the ions are ejected and transferred to the hyperbolic Penning trap where the mass measurement is carried out using a time-of-flight resonance technique [16, 17]. The mass measurement is performed by a determination of the cyclotron frequency $\nu_c = qB/(2\pi m)$ of stored ions with mass m and charge q in a homogeneous magnetic field B . The time of flight of the ions from the trap to a micro-channel-plate detector (MCP5 in Fig. 1) is recorded after probing the ions with an RF-excitation frequency and ejecting them. Repeating this for different RF-excitation frequencies and measuring the time of flight as a function of the radiofrequency, a time-of-flight cyclotron resonance curve is obtained (see inset in Fig. 1). The frequency of the absolute minimum of this curve is the cyclotron frequency of the stored ions.

The calibration of the magnetic field B is performed by a determination of the cyclotron frequency of a reference ion ν_c^{ref} with well-known mass, before and after the measurements of the cyclotron frequency of the ion of interest ν_c . The value adopted for ν_c^{ref} is the result of the linear interpolation of both measurements (before and after) to the center of the time interval during which the cyclotron frequency of the ion of interest was measured. In that way, a possible linear shift of the magnetic field is accounted for. However, besides the measured long-term drifts of the magnetic field, there are short-term fluctuations of B . These were measured using $^{85}\text{Rb}^+$ during a period of 300 hours at ISOLTRAP [18]. The results lead to an additional relative uncertainty of the cyclotron frequency of the ions of reference, of $u_{\text{mag}}(\nu_c^{\text{ref}})/\nu_c^{\text{ref}} = 3.8 \times 10^{-9} / \text{h}$ [18]. This uncertainty is quadratically added to the uncertainty of the cyclotron frequency of the reference ion.

The presence of contaminating ions in the measurement trap, produced either in the ISOLDE plasma ion source or created by charge exchange in the ISOLTRAP preparation traps, induces a shift in the cyclotron frequency of the ion of interest due to the coupling of

the eigenmotions brought about by Coulomb interaction between ions with different mass-to-charge ratio [19]. This shift can be corrected for by applying a count rate analysis. In this analysis, the cyclotron frequency is plotted as a function of the number of ions stored simultaneously in the trap, or more precisely, the number of detected ions per measurement cycle, i.e. trap filling. The true cyclotron frequency is then determined by linear extrapolation to one ion stored at a time. The extrapolation is done to 0.25 detected ions, assuming a detector efficiency of about 25%.

Two more uncertainties are quadratically added to the uncertainty of the resulting cyclotron frequency ratio. These are the mass-dependent and the residual systematic uncertainties. The magnitudes of both are results from cross-reference measurements carried out with carbon clusters [18, 20]. The mass-dependent uncertainty is a function of the mass difference between the reference ion and the ion of interest and reads $u_m(r)/r = 1.6 \cdot 10^{-10}/u \cdot (m - m_{\text{ref}})$, with u being the atomic mass unit. The residual relative systematic uncertainty also obtained from these measurements is 8×10^{-9} .

The atomic mass is determined from the measured ion cyclotron frequencies via the relationship

$$m_{\text{atom}} = r \cdot (m_{\text{atom}}^{\text{ref}} - m_e) + m_e, \quad (1)$$

where r is the cyclotron frequency ratio between the reference ion and the ion of interest obtained in the experiment. m_e is the electron mass and $(m_{\text{atom}}^{\text{ref}} - m_e)$ is the reference ion mass, here $^{85}\text{Rb}^+$.

III. EXPERIMENT

The data presented in this paper were collected during two data taking periods, one in October 2000 (Run 1) and another in September 2001 (Run 2). Each period lasted for about one week and a total of 74 hours of data were recorded. The radioactive krypton isotopes were produced in spallation reactions as a result of bombarding a ZrO_2 ($\rho = 8 \text{ g/cm}^2$), and a Nb foil target ($\rho = 50 \text{ g/cm}^2$), with the intense proton beam from the CERN PS-Booster accelerator [21]. $3.2 \cdot 10^{13}$ protons with an energy of 1.4 GeV impinged on the target every 2.4 s. Besides the nuclides produced in the target, other elements present in the construction material of the plasma ion source can be ionized. These are, for example, Ta from the cathode, C from the grids, Mo contained in the plasma chamber, or borium

nitride from the insulators [22]. In addition to these elements, oxygen is always present leading to the formation of oxide molecules like CO, CO₂, TaO etc.. In order to reduce these contaminants, a water-cooled transfer line was used between target and ion source. In that way, only the volatile products (noble gases, O₂, N₂, etc.) and occasionally Rb and Br are transported into the plasma ion source biased at +60 kV. The stable isotopes of Kr were produced by leaking Kr gas into an ion source. The separator in use was the HRS.

The accumulation time in the RFQ buncher was varied, depending on the krypton isotope, between 20 μ s (for ⁸⁶Kr) up to 50 ms (for ⁷²Kr). The time for purification in the gas-filled cylindrical Penning trap was 125 ms. The limitation in the time for preparation was imposed by the expected losses due to charge exchange which occur by collisions with impurities in the buffer gas. In the precision Penning trap, the radioactive krypton isotopes were measured by using excitation times (T_{RF}) of 300 ms, 400 ms and occasionally 900 ms. The cyclotron frequency line-width $\Delta\nu_c$ (FWHM) is inversely proportional to the excitation time, thus resulting in resolving powers $m/\Delta m$ (FWHM) of at least 4×10^5 for ^{72–77}Kr⁺, in the 5.9 T strong homogeneous magnetic field of the ISOLTRAP precision Penning trap. The excitation time for the stable reference ions ⁸⁵Rb⁺ produced by an internal ion source was $T_{\text{RF}} = 1.2$ s. The mass of ⁸⁵Rb⁺ has been very precisely determined with a relative mass uncertainty of 2×10^{-10} in a Penning trap experiment at MIT [23]. The period between the reference measurements (before and after) was varied from thirty minutes in the case of ⁸⁶Kr⁺ up to six hours in the case of ⁷²Kr⁺. This time was closely related to the production yield and, in the particular case of Kr isotopes, to strong ion losses due to charge exchange.

In most of the measurements reported here, only one or very few ions were simultaneously confined in the precision Penning trap and the cyclotron frequency obtained from the count-rate analysis of ions overlaps with that without separating the number of detected ions into classes. In those cases, the count rate analysis leads only to a larger uncertainty in the determination of the cyclotron frequency. In some cases, like for ⁷²Kr, most of the events recorded correspond to one ion per proton pulse so that correction for contamination is not practicable.

The cyclotron frequency ratios and the resulting mass values according to Eq. 1 are given in Tab. I.

IV. ATOMIC-MASS EVALUATION

The mass excess D of a nucleus is given by

$$D = m_{\text{atomic}} - A \cdot u, \quad (2)$$

where A is the atomic mass number of the nucleus and u the atomic mass unit, defined as $1/12$ of the mass of ^{12}C . The mass excesses of all known nuclides are tabulated in the Atomic-Mass Evaluation (AME), which takes all available experimental data as input [28]. Each nuclide can be linked to up to four other nuclides. The adjustment procedure is performed using the least-squares method for all nuclides having more than one link to other nuclides (primary nuclides). The masses of the primary nuclides are adjusted and subsequently used for the mass determination of nuclides with only one link to other nuclides (secondary nuclides).

In the case of several input data for the same link between two nuclides, the input will be the weighted mean of such data. If the uncertainties differ by more than a factor of three, only the most precise value will be considered since the contributions from the other values will be almost negligible. In the case of discrepancies, the way in which the measurement was performed (described in the original publication) will be re-examined in order to accept or reject an experimental datum. A detailed description of the procedure is given in Ref. [28].

Table II shows the mass excess values resulting directly from our measurements of the cyclotron frequencies, from the AME prior to the ISOLTRAP measurement [27] and those including our data. The column $D_{\text{exp}} - D_{\text{pre}}$ in Tab. II gives the differences between the mass excess values (D_{exp}) resulting from the measurements reported here and the previous mass excess values (D_{pre}) from AME'95 [27]. Figure 2 shows these deviations for the nuclides $^{72-78,80,82}\text{Kr}$. The results are discussed in detail in the following.

^{72}Kr

The $N = Z$ nuclide ^{72}Kr was previously determined from the Q -value of the β^+ -decay to ^{72}Br [30] with a relative mass uncertainty of $\delta m/m = 1.8 \cdot 10^{-6}$. The nuclide was produced in the $^{58}\text{Ni}(^{16}\text{O}, 2n)^{72}\text{Kr}$ reaction. The measured Q -value in this experiment was 5057(135) keV and the mass excess $D(^{72}\text{Kr}) = -54110(270)$ keV [27]. The ISOLTRAP value for ^{72}Kr is $D(^{72}\text{Kr}) = -53940.6(8.0)$ keV. This value is in agreement with the previous one but has a relative mass uncertainty of $u(m)/m = 1.2 \cdot 10^{-7}$. Thus, the new AME value is determined to 100% by the ISOLTRAP result.

⁷³Kr

The prior mass excess value of ⁷³Kr was determined by two experiments, one using the ⁷³Kr(β^+)⁷³Br decay [30] and the other the β^+ -delayed proton decay of ⁷³Kr [31] (see Fig. 3). In the work of Schmeing *et al.*, ⁷³Kr was produced as a by-product in the ⁵⁸Ni(¹⁶O, n)⁷³Kr reaction and the β end point energy was measured in a β - γ coincidence measurement. From this experiment, a Q -value of $Q_{\text{EC}} = 6790(350)$ keV yielded a mass excess of $D(^{73}\text{Kr}) = -56775(350)$ keV. In the work of Hardy *et al.*, ⁷³Kr was produced in the reaction ⁶⁰Ni(¹⁶O, $3n$)⁷³Kr. They recorded the energy spectrum of protons emitted from the intermediate highly excited states of ⁷³Br. The proton end point energy then yielded a decay energy $Q_{\text{EC}} - B_p = 3700(150)$ keV, corresponding to a mass excess of $D(^{73}\text{Kr}) = -56905(150)$ keV. The AME value based on these measurements was $-56890(140)$ keV, from which the ISOLTRAP value $-56551.7(6.6)$ keV differs by 340 keV. Note that this value is the outcome of two different ISOLTRAP experiments yielding $D = -56550.8(9.0)$ keV and $D = -56552.7(9.5)$ keV, respectively. The ISOLTRAP value is in agreement with the one from Schmeing *et al.*, and deviates by 2.3σ from that of Hardy *et al.* Though larger than expected, this disagreement can be due to deformation of this nucleus, which can affect the distribution of β feeding and distort the spectrum in a way that might lead to a shift in the fitted proton spectrum shape and, hence, the end point [32]. This hypothesis is supported by the fact that the measured proton branching ratio strongly exceeded that from statistical model calculations. Thus, a second value of $Q_{\text{EC}} - B_p$, less sensitive to nuclear structure, was reported in Ref. [31]. They assumed that all the protons directly populated the ground state of ⁷²Se. They measured the coincidences between protons and 511 keV γ -rays at the corresponding proton energy, and accounted for the proton branching ratio to the first excited state of ⁷²Se by their statistical model calculations. In this case, the measured proton branching ratio to the first excited state of ⁷²Se (18(4)%) was in agreement with the statistical model calculations (18% assuming $J^\pi = 5/2^+$). The value obtained that way was less precise ($Q_{\text{EC}} - B_p = 3900(400)$ keV), but the mass value agrees better with our measurement. This value and the one from Schmeing *et al.* are outweighed and the data from the two ISOLTRAP experiments now completely determine the adjusted value.

⁷⁴Kr

The ISOLTRAP data for the mass of ⁷⁴Kr deviate by 2.7σ from the previous AME value,

which was determined from three measurements [33–35] (see Fig. 4). The first two mass excess values were obtained from $\beta^+-\gamma$ coincidence measurements. As shown in Fig. 4, the most precise of these two values [34] deviates by 2.8σ from the result presented in this paper. The deviation can be assigned to the so-called “Pandemonium” effect [32]. This effect was reported for the first time by Hardy *et al.* [36]. It appears in cases where the Q -value is high, such that there can be many weak β transitions to highly excited states that subsequently generate a large number of weak γ -ray transitions. A significant fraction of these transitions will be unobserved individually but will collectively populate low-lying states to an appreciable extent, affecting the distribution of β feeding and therefore the spectrum obtained from the coincidence measurement. The last and most precise of the three values was obtained from the determination of the Q -value of the $^{78}\text{Kr}(^4\text{He}, ^8\text{He})^{74}\text{Kr}$ reaction by Moltz *et al.* [35]. This result shows a 1.4σ deviation from the ISOLTRAP mass excess, even after taking into account the change in the ^{78}Kr mass due to our new measurement. The energy calibration in that experiment was done through the $^{64}\text{Ni}(^4\text{He}, ^8\text{He})^{60}\text{Ni}$ reaction. The reaction Q -value was extracted from a gated ^8He position spectrum. In this case, due to the poor statistics, the extracted Q -value depends on the number of counts identified as stemming from the reaction. Considering the much smaller uncertainty of the ISOLTRAP result and the good agreement between the two ISOLTRAP experiments ($D(^{74}\text{Kr}) = -62330.3(2.4)$ keV and $D(^{74}\text{Kr}) = -62336.9(4.1)$ keV), all other measurements are outweighed.

^{75}Kr

The previous value for the mass excess of ^{75}Kr was determined by one measurement, using the $^{78}\text{Kr}(^3\text{He}, ^6\text{He})^{75}\text{Kr}$ reaction [37]. The mass excess, with an uncertainty of about three times the uncertainty of the ISOLTRAP value, deviates by 4.5σ from our value. In Ref. [37] a 70-MeV ^3He beam was directed onto an enriched ^{78}Kr gas target. The position spectrum of the reaction product ^6He at a scattering angle of $\theta_{\text{lab}} = 7.25^\circ$ was recorded at the focal plane of a split-pole spectrograph. The reaction $^{18}\text{O}(^3\text{He}, ^6\text{He})^{15}\text{O}$ was used for the calibration of the detector. Three peaks were identified as representing the population of the $7/2^-$ state at 611 keV, the $5/2^-$ at 358 keV, and the $5/2^+$ ground state, respectively. The assignment appears to be correct, but the reported uncertainty of 14 keV may have been underestimated considering that it corresponds to only one half channel. The analogous measurement of the reaction energy of the $^{82}\text{Kr}(^3\text{He}, ^6\text{He})^{79}\text{Kr}$ reaction agrees well with the expected value. The third analogous measurement, that of the $^{80}\text{Kr}(^3\text{He}, ^6\text{He})^{77}\text{Kr}$ reaction,

also deviates by several standard deviations from the current value. This supports the assumption that the uncertainties were underestimated. Interestingly, although the statistics seem to be very similar for all three measurements, the uncertainty assigned to the Q value of the $^{78}\text{Kr}(^3\text{He},^6\text{He})^{75}\text{Kr}$ reaction is much smaller than that of the two other measurements. Thus, the datum is excluded from the adjustment. The influence of the ISOLTRAP datum is then 100%.

⁷⁶Kr

The mass of ^{76}Kr was determined to 31 % by a reaction energy measurement of $^{78}\text{Kr}(\text{p,t})^{76}\text{Kr}$ [38] and to 69 % from the $^{78}\text{Kr}(^4\text{He},^6\text{He})^{76}\text{Kr}$ reaction Q value [35]. These two measurements have a very good consistency of $\pm 0.1\sigma$. The new ISOLTRAP value is in agreement within 1σ with the data from Matsuki *et al.* [38], but deviates by 1.4σ from the original datum from Moltz *et al.* [35]. A closer study of the paper revealed that the Q value was in fact measured on a relative scale, with respect to that of the $^{80}\text{Kr}(^4\text{He},^6\text{He})^{78}\text{Kr}$ reaction. This means that the measurement really connects the three masses of ^{80}Kr , ^{78}Kr , and ^{76}Kr . The Q -values of the two ($^4\text{He},^6\text{He}$) reactions are:

$$Q_1 = D(^{80}\text{Kr}) - D(^{78}\text{Kr}) + D(^4\text{He}) - D(^6\text{He}) \quad (3)$$

$$Q_2 = D(^{78}\text{Kr}) - D(^{76}\text{Kr}) + D(^4\text{He}) - D(^6\text{He}). \quad (4)$$

The measured quantity is $\Delta Q = Q_1 - Q_2$, thus:

$$\Delta Q = D(^{80}\text{Kr}) + D(^{76}\text{Kr}) - 2D(^{78}\text{Kr}). \quad (5)$$

This value is not given in the paper, but using the reported value $Q_2 = -20351(10)$ keV and the mass excesses of ^{80}Kr , ^{78}Kr , ^6He , and ^4He available in the year 1978 [39], one can obtain $\Delta Q = 1432$ keV. The AME datum for this reaction was therefore changed to reflect the relation of Eq. (5) and assigned a value of $1432(10)$ keV. This replacement, however, does not remove the disagreement with the ISOLTRAP datum. Only the most precise of the two previously used measurements, *i.e.* the value from Moltz *et al.* [35], is retained as an input parameter for the mass evaluation together with the new ISOLTRAP value.

⁷⁷Kr

Before the new ISOLTRAP measurement, the mass of ^{77}Kr was determined by two different measurements made by the same group [37]. The more precise one was obtained from the $^{78}\text{Kr}(d,t)^{77}\text{Kr}$ reaction Q value. The other one was obtained by a measurement similar to the $(^3\text{He},^6\text{He})$ reaction described above for ^{75}Kr . The two data are in good agreement with each other. An older value from a β end point measurement [40] deviates from their average by 1.9σ . The ISOLTRAP value is in agreement with the result from the $(^3\text{He},^6\text{He})$ reaction from Moltz *et al.* [37], but the (d,t) Q value deviates from the ISOLTRAP value by 2.5σ . Moltz *et al.* [37] bombarded a ^{78}Kr gas target with a 29.1-MeV deuteron beam and observed the triton position spectrum in a split-pole spectrograph. The focal plane was calibrated with tritons from the $^{22}\text{Ne}(d,t)^{21}\text{Ne}^*$ (351 keV) and the $^{40}\text{Ar}(d,t)^{39}\text{Ar}^*$ (1267, 1517, and 2358 keV) reactions. The smallest uncertainty is the one from the (d,t) reaction, which retains some weight while the value from the $(^3\text{He},^6\text{He})$ reaction is outweighed. In Thulin's paper, only a β -end-point energy of 1860 keV without an uncertainty is given, suggesting that the decay populates the ground state of ^{77}Br . When the datum was included in the AME, it was re-interpreted as populating the 129.6 keV level and was assigned an uncertainty of 30 keV. Considering the complexity of the Fermi-Kurie analysis that led to the end point energy, the uncertainty of the energy was possibly underestimated. In the new AME the contribution of the ISOLTRAP value is 93%.

^{78}Kr

^{78}Kr is a stable isotope with an isotopic abundance of 0.35%. Its mass was measured for the first time in 1963 with a double-focusing mass spectrometer [41] and its mass excess was determined to be $D(^{78}\text{Kr}) = -74180.5(3.4)$ keV. This value was incorrectly transcribed into the AME'93 as $-74147.0(3.4)$ keV. Later mass measurements were carried out by the determination of the reaction energy of $^{78}\text{Kr}(d,t)^{77}\text{Kr}$ and $^{78}\text{Kr}(^3\text{He},^6\text{He})^{75}\text{Kr}$ [37], and by the reaction $^{78}\text{Kr}(^3\text{He},d)^{79}\text{Rb}$ [42]. All the results are shown in Fig. 5. In the original publications, the reactions $^{78}\text{Kr}(d,t)^{77}\text{Kr}$, $^{78}\text{Kr}(^3\text{He},^6\text{He})^{75}\text{Kr}$, and $^{78}\text{Kr}(^3\text{He},d)^{79}\text{Rb}$ were used to determine the masses of ^{77}Kr , ^{75}Kr and ^{79}Rb from the mass of ^{78}Kr . The determination of the mass of ^{78}Kr via this method requires the knowledge of the masses that are linked through Q -values to this primary nuclide. Using the masses of the former three as given in AME'95, weighted with their uncertainties, yielded a mass excess of $-74160(7)$ keV. The mass excess measured at ISOLTRAP in the first experiment was $-74179.2(1.3)$, which is in agreement with $D = -74182.4(2.2)$ obtained from the second experiment. The ISOLTRAP

mass agrees well with the result from Ries *et al.* [41] after correction, and with the result from Stephans *et al.* [42]. The uncertainty in the result from Stephans *et al.* is outweighed by the value from Ries *et al.* and by our datum. The contribution of the ISOLTRAP value to the final value is 93 %.

⁸⁰Kr

⁸⁰Kr is also a stable isotope with an isotopic abundance of 2.25%. Our new datum for the mass of ⁸⁰Kr agrees well with the previously accepted value. The six measurements that contribute to the accepted value in AME'95 agree very well with each other with a maximum deviation of 0.6σ . In particular, the three most precise previous determinations agree well with the ISOLTRAP result and will therefore not be discussed in detail. The Q -value of the ⁸⁰Kr(³He, ⁶He)⁷⁷Kr reaction from Moltz *et al.* [37], being the least precise of all measurements, is removed from the adjustment calculation of the AME, all others remain included. An additional datum due to the modified (³He, ⁶He) reaction value from Moltz *et al.* is newly introduced. Its disagreement with our result is of course the same (in opposite direction) as that for the mass of ⁷⁶Kr and has already been discussed there. The influence of the ISOLTRAP result on ⁸⁰Kr is 86% in the AME2003.

⁸²Kr

The natural isotopic abundance of the stable isotope ⁸²Kr is 11.6%. The AME'95 mass value of ⁸²Kr has contributions from six different measurements using a wide range of techniques and mass relations. They agree well with each other (within $\leq 0.9\sigma$). The new ISOLTRAP mass value with a slightly smaller uncertainty than that of the previous measurements deviates by 0.7σ from the AME'95 value, but the three most precise of the previous measurements, *i.e.*, the result from a β -end-point determination of the ⁸²Kr(β^-)⁸²Br decay [43], a mass-spectrometry measurement [41], and the fairly recent datum from a high-resolution deflection mass spectrometer [44] are in better agreement with our value. For the adjustment, all old input parameters are kept and the ISOLTRAP datum is added. It influences the final result by 48%.

⁸⁶Kr

⁸⁶Kr is the second most abundant naturally occurring krypton isotope (17.3%). Its mass was measured with an uncertainty of only 0.1 keV by SMILETRAP, [$D(^{86}\text{Kr}) = -83265.4(1)$ keV] [45]. Our value [$D(^{86}\text{Kr}) = -83265.5(3.3)$ keV] is in excellent agreement with the SMILETRAP measurement but a factor of 30 less precise. Therefore it is not

included in the atomic mass evaluation.

V. CONCLUSION

In this paper, the results from two ISOLTRAP experiments performed in 2000 and in 2001 were presented. The masses of $^{72-78,80,82}\text{Kr}$ were measured directly with unprecedented precision and the influence of these measurements in the AME was discussed. For the most exotic Kr isotopes ($^{72-74}\text{Kr}$), the uncertainties on the mass values were improved by at least a factor of 20. These mass values have already been used for astrophysical calculations and tests of the SM. The nuclide ^{72}Kr has been shown to be a strong waiting point in the rp -process delaying the Type I X ray burst duration by at least 80% of its β -decay lifetime [6]. The Ft -value for the transition $^{74}\text{Rb}(\beta^+)^{74}\text{Kr}$ has been determined very accurately yielding an accurate isospin-symmetry-breaking parameter [9]. This is important since strong deviations are observed among different theoretical calculations of this parameter in this nuclear shell region.

The ISOLTRAP values on $^{73,74}\text{Kr}$ deviate considerably from the previous results obtained using indirect techniques. These results date back from the seventies and eighties and the methods in use at that time might not be reliable for these particular Kr isotopes. The mass of ^{73}Kr was determined from the end point energy of the protons emitted from the intermediate highly excited state of the daughter nuclei, and from the Q -value of the β^+ transition. ^{73}Kr shows strong nuclear deformation and the measured proton end point energy is sensitive to nuclear structure effects. The Q -value was obtained from a β - γ coincidence measurement which is not suitable in the case of exotic nuclei with large Q -values due to the “Pandemonium” effect. This effect leads, in general, to Q -values that are too low. This was not observed for ^{73}Kr due to the large uncertainty of the reported value, but can explain the deviation observed for ^{74}Kr . Other measurements on ^{74}Kr were revised and are in agreement with the ISOLTRAP results. However, the much improved precision in the ISOLTRAP results, a factor of 21 for ^{73}Kr and a factor of about 29 for ^{74}Kr , set them as the new AME values. The difference between the ISOLTRAP mass value on ^{72}Kr and the previous one was smaller than its quoted uncertainty which is now reduced by a factor of about 34. The previous mass AME value of ^{75}Kr , measured indirectly, had an uncertainty of only 15 keV but differed by more than 5σ from the ISOLTRAP value. The uncertainty

of 15 keV seems to be underestimated on comparing it with that of other measurements which apply the same technique. Thus, the previous mass value has been substituted by the ISOLTRAP result.

For the other Kr isotopes $^{76-78,80,82,86}\text{Kr}$ (long-lived or stable), the influence of the ISOLTRAP results is smaller than 100%, *i.e.*, 85% for ^{76}Kr , 93% for ^{77}Kr and ^{78}Kr , 86% for ^{80}Kr and 48% for ^{82}Kr . In the case of ^{76}Kr and ^{78}Kr , the differences between the old and the new AME results were of a few tens of keV. The mass of ^{86}Kr , though measured along with the other masses reported here, had no influence on the AME2003 since a compatible result but with a factor of 33 higher precision was obtained at SMILETRAP using highly-charged ions. The agreement between the results from ISOLTRAP and SMILETRAP supports the validity of our other measurements.

VI. ACKNOWLEDGEMENT

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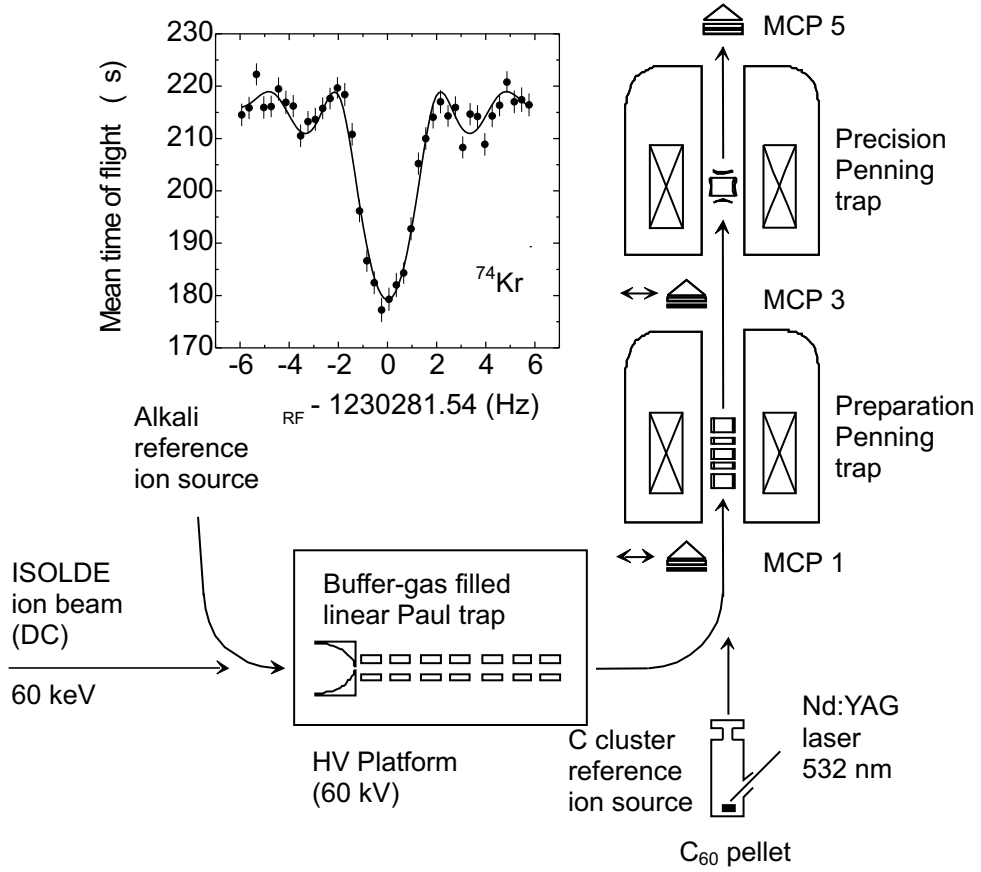


FIG. 1: Sketch of the ISOLTRAP setup. It consists of three traps, a buffer-gas-filled linear Paul trap, and two Penning traps housed in different superconducting magnets. In addition, an off-line ion source supplies the stable reference ions. In the experiments reported here, an alkali ion source was used for that purpose. The carbon cluster ion source, also shown in the sketch, has been extensively tested and will be implemented in the future to replace the alkali ion source for absolute mass measurements using carbon clusters C_n ($1 \leq n \leq 22$) as reference ions. The time-of-flight cyclotron resonance is measured using the micro-channel plate detector MCP5. MCP1 and MCP3 are micro-channel plate detectors used for diagnosis.

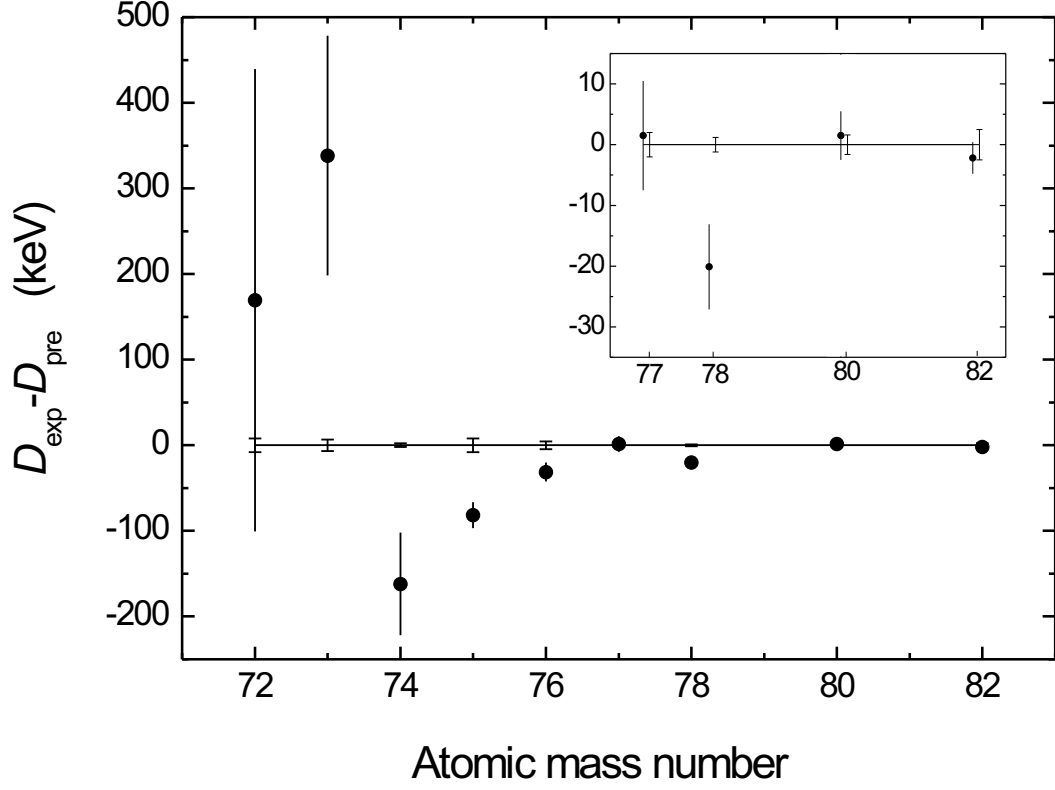


FIG. 2: Differences between mass excesses of Kr isotopes as determined in this work and from the AME'95 [27]. The zero line is defined by the ISOLTRAP results (D_{exp} from Tab. II), the uncertainties of which are indicated by the symbol I. The AME'95 values (D_{pre} from Tab. II) are indicated by full dots. The inset is an enlargement of the mass range 77-82. The ISOLTRAP results are displayed to the right of the AME'95 values to allow for a clear view.

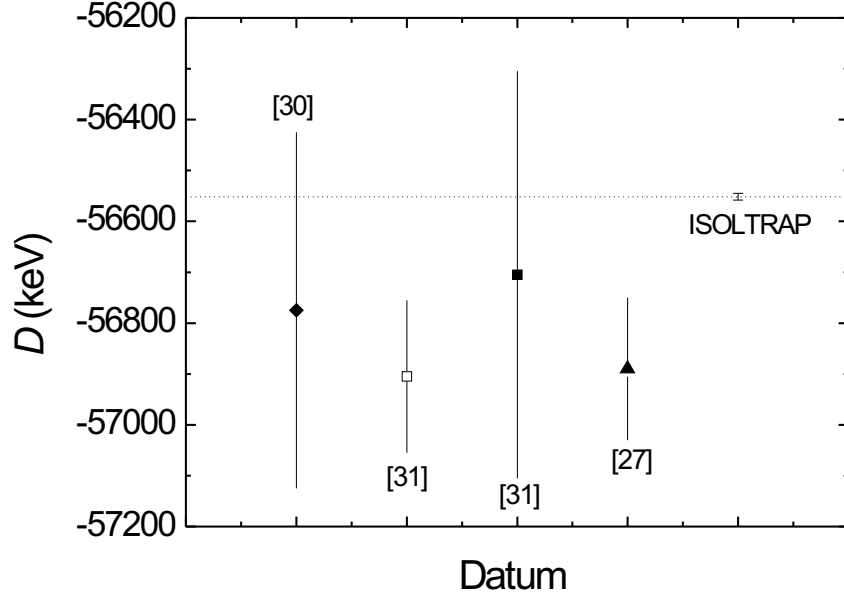


FIG. 3: Comparison between the ISOLTRAP result (dotted line) and the previous mass excess values for ^{73}Kr . The two previous mass values, represented by the diamond and the empty square in the figure, were obtained, respectively, from the Q -value, and from the energy spectrum of protons emitted from the intermediate highly excited states of the daughter nuclei. They determined the AME value in 1995. The filled square is the mass excess value based on an evaluation which is less sensitive to nuclear structure than the one yielding the empty square. The result of this measurement was also reported by Hardy *et al.* [31]. See text for further details.

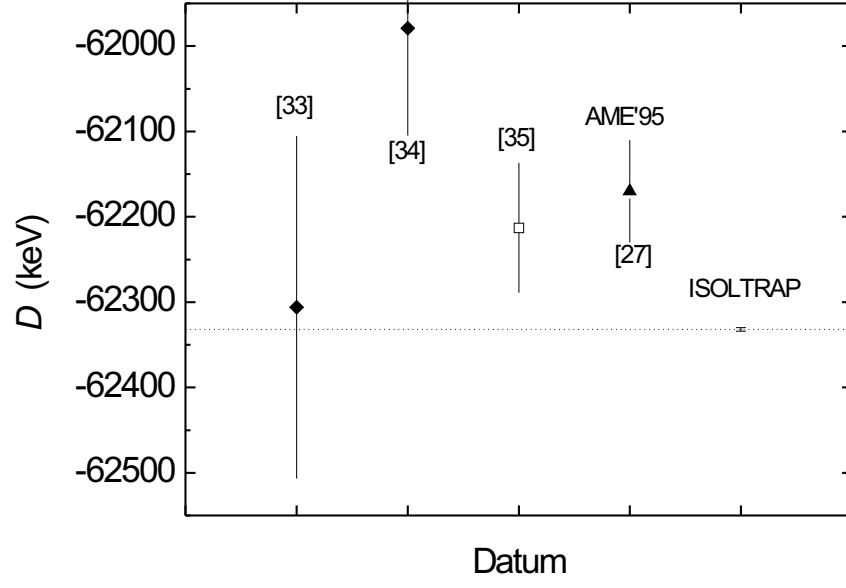


FIG. 4: Comparison between the ISOLTRAP result (dotted line) and the previous mass excess values for ^{74}Kr . The three previous mass values, represented by the diamonds and the empty square in the figure, were obtained from reaction energies and determined the AME value in 1995.

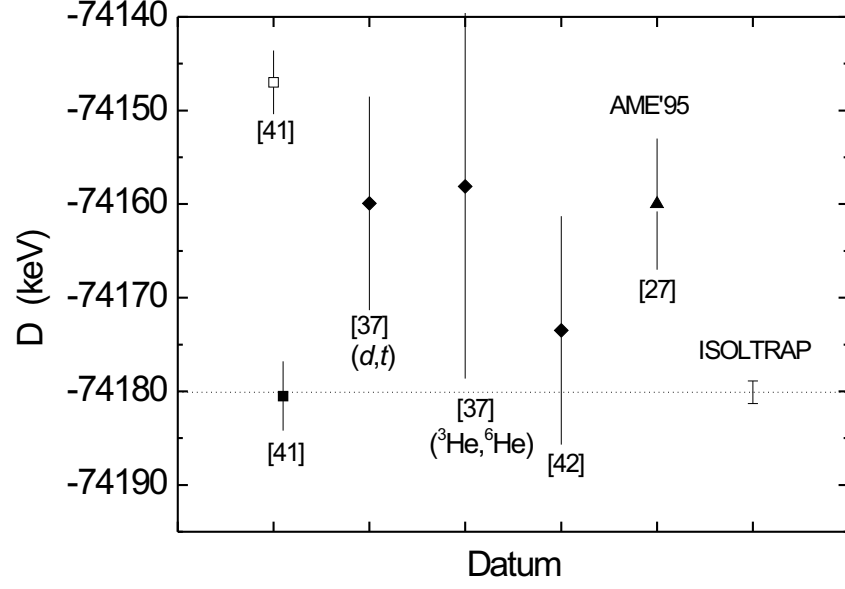


FIG. 5: Comparison between the ISOLTRAP mass excess value (dotted line) and the previous results for ^{78}Kr . The AME'95 value was determined using the mass excess values represented in the figure by the diamonds and the empty square. This mass excess value (empty square) represents a datum that was incorrectly transcribed into the AME'93. After correction, the mass excess value becomes that represented by the filled square.

TABLE I: Frequency ratios ν_c^{ref}/ν_c relative to $^{85}\text{Rb}^+$. The uncertainties of the frequency ratios are given with three digits in order to reduce roundings errors. m_{atom} represents the atomic mass calculated according to Eq. (1) taking $m(^{85}\text{Rb}) = 84.911\,789\,738(12)$ u [23], and $m_e = 0.000\,548\,579\,9110(12)$ u [24]. $[u(m)/m]_{\text{atom}}$ gives the relative mass uncertainty. The half-lives $T_{1/2}$ are taken from [25] and N is the number of detected ions. The excitation times T_{RF} given in the table were the most commonly used in the experiments.

Nuclide	$T_{1/2}$	Run	N	T_{RF} (ms)	ν_c^{ref}/ν_c	m_{atom} (u)	$[u(m)/m]_{\text{atom}}$ (10^{-7})
^{72}Kr	17.2(3) s	2	446	400	0.847 255 827(101)	71.942 0924(86)	1.2
^{73}Kr	27.0(1.2) s	1	107	300	0.858 999 829(114)	72.939 2902(97)	1.3
^{73}Kr	,,	2	2850	400	0.858 999 804(121)	72.939 2881(102)	1.4
^{74}Kr	11.50 min	1	5649	300, 900	0.870 703 7618(303)	73.933 0857(26)	0.3
^{74}Kr	,,	2	21304	400	0.870 703 6780(516)	73.933 0785(44)	0.6
^{75}Kr	4.29 min	1	1492	300	0.882 455 563(102)	74.930 9457(86)	1.2
^{76}Kr	14.8 h	1	3433	300	0.894 173 3117(557)	75.925 9143(47)	0.6
^{77}Kr	74.4 min	1	35639	300	0.905 935 6611(248)	76.924 6700(21)	0.3
^{78}Kr	<i>stable</i>	1	64604	300	0.917 661 9688(159)	77.920 3653(13)	0.2
^{78}Kr	,,	2	40705	400	0.917 661 9283(254)	77.920 3619(22)	0.3
^{80}Kr	<i>stable</i>	1	11510	1400	0.941 169 0366(199)	79.916 3796(17)	0.2
^{82}Kr	<i>stable</i>	1	9032	900	0.964 688 9212(305)	81.913 4822(27)	0.3
^{86}Kr	<i>stable</i>	2	17201	600	1.011 763 1202(421)	85.910 6108(36)	0.4

TABLE II: Results of the Atomic Mass Evaluation. D_{exp} represents the ISOLTRAP experimental mass excess obtained from the cyclotron frequency ratios given in Tab. I with $1 \text{ u} = 931494.009(7) \text{ keV}$ [29]. D_{pre} are the AME values from 1995 [27], and D_{new} are the new AME results including D_{exp} . The last column gives the influence of the ISOLTRAP data on the AME2003 values.

Nuclide	D_{exp} (keV)	D_{pre} (keV)	D_{new} (keV)	$D_{\text{exp}} - D_{\text{pre}}$ (keV)	Influence (%)
^{72}Kr	-53940.6(8.0)	-54110(270)	-53940.6(8.0)	169.4	100
^{73}Kr	-56551.7(6.6)	-56890(140)	-56551.7(6.6)	338.3	100
^{74}Kr	-62332.0(2.1)	-62170(60)	-62332.0(2.1)	-162.0	100
^{75}Kr	-64323.6(8.0)	-64242(15)	-64323.6(8.1)	-81.6	100
^{76}Kr	-69010.4(4.4)	-68979(11)	-69013.8(4.0)	-31.4	85
^{77}Kr	-70169.5(2.0)	-70171(9)	-70170.8(1.9)	1.5	93
^{78}Kr	-74180.1(1.2)	-74160(7)	-74179.3(1.1)	-20.1	93
^{80}Kr	-77891.9(1.6)	-77893.4(4)	-77892.3(1.3)	1.5	86
^{82}Kr	-80590.8(2.5)	-80588.6(2.6)	-80589.6(1.7)	-2.2	48
^{86}Kr	-83265.5(3.3)	-83265.9(1.1)	-83265.4(0.1)	0.4	0